Trends of OCS, HCN, SF<sub>8</sub>, CHCIF<sub>2</sub> (HCFC-22) in the Lower Stratosphere from 1985 and 1994 Atmospheric Trace Molecule Spectroscopy Experiment Measurements near 30°N Latitude

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Abstract. Volume mixing ratio (VMR) profiles of OCS, HCN, SF<sub>6</sub>, and CHCIF<sub>2</sub> (HCFC-22) were measured near 30°N latitude by the Atmospheric Molecule Spectroscopy Fourier transform spectrometer during shuttle flights on 29 April-6 May 1985 and 3-12 November 1994. The change in the concentration of each molecule in the lower stratosphere has been derived for this 94-year period by comparing measurements between potential temperature of 395 to 800 K (~17 to 30 km altitude) relative to simultaneously measured values of the long-lived tracer  $N_2O$  , Ratios of the 1994-to-1985 VMRs and the corresponding exponential increase rates inferred from these comparisons are  $0.94\pm0.03$  and  $(-0.6\pm0.3)$ % yr<sup>-1</sup> for OCS, 1.09±0.09 and (0.9±0.9) yr<sup>-1</sup> for HCN, 1.96\*0.06 and (7.1±0.3) yr<sup>-1</sup> for SF, 2.08 $\pm$ 0.24 and (7.7 $\pm$ 1.2)% yr<sup>-1</sup> for CHC&F<sub>2</sub> (HCFC-22), 1 sigma. These results have been compared with trends reported in the literature.

#### Introduction

The Atmospharic Trace Molecule Spectroscopy (ATMOS) instrument is a 0.01-cm<sup>-1</sup> resolution Fourier transform spectrometer (FTS) designed to record broadband, midinfrared solar occultation

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spectra of the middle atmosphere from low Earth orbit [Farmer, 1987], To date, the ATMOS FTS has flown 4 times onboard the shuttle as part of the Spacelab 3 (29 April-6 May 1985), ATLAS (Atmospheric Laboratory for Applications and Science) 1 (24 March-2 April 1992), ATLAS 2 (8.17 April 1993), and ATLAS 3 (3-12 November 1994) missions. Gunson et al. [1996] have provided a summary of the scientific objectives of the ATMOS experiment and a plot of the latitudinal and longitudinal coverage of the observations recorded during each flight,

An important overlap in coverage occurred at 28°N-33°N latitude where xx and xx occultations were recorded at sunset during the Spacelab 3 and ATLAS 3 flights, respectively. The middle atmosphere profiles of more than 2 dozen molecules derived from these measurements provide a unique record of changes in stratospheric composition at northern lower mid-latitudes over the 94 years separating the 2 missions. An evaluation of changes in stratospheric chlorine loading [Zander et a]., 1996a] and stratospheric CF, has been reported on the basis of these observations [Zander et al., 1996b],

The purpose of the present Letteris to use the Spacelab 3 and ATLAS 3 measurements in this common latitudinal band to assess changes in the volume mixing ratios (VMRs) of 4 important trace OCS, HCN, SFa, and CHCIF2 molecules: (HCFC-22). For each molecule, measured VMRs in the lower stratosphere are referenced to simultaneous values for conserved, long-lived dynamical tracer N20 to remove the contribution of atmospheric dynamicals to the observed variations of the target molecules, 1985-1994 trends inferred from the VMR difference have been compared with values reported in the literature for the same time period.

### Spectroscopic Analysis

The pressure-temperature and constituent profiles were derived from

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the ATMOS spectra with the Occultation Spectra (ODS) onion-peeling Display retrieval Igorithm [Norton and Rinsland, 1991] and the 1995 ATMOS line parameters compilation [Brown et al., 1996], that except for HCN, the spectroscopic parameters for the target molecules were significantly changed as compared with those on the previous version of the ATMOS database [Brown et al., 1987]. A synopsis of the procedure used to produce the version 2 VMR profiles reported in this paper has been provided by Abrams et al, [1996]. The CO<sub>2</sub> profile is a fundamental assumption in the ATMOS analysis procedure as it is the basis for the pressure-temperature subsequent VMR retrievals, The CO2 VMRs assumed for the Spacelab 3 mission corresponds to a decreasing from  $3,40 \times 10^{-4}$  at the surface to  $3,33 \times 10^{-4}$  in the middle and upper stratosphere. corresponding values for the ATLAS 3 mission are  $3,64 \times 10^{-4}$  and  $3,51 \times 10^{-4}$ , respectively,

For OCS and HCN, narrow intervals ("microwindows") containing isolated, unblended spectral features were fitted by line-by-line methods over prespecified ranges in tangent altitude, A weighted mean and weighted standard deviation of individual profiles were calculated foreach occultation and interpolated to a potential temperature (0) grid. Six OCS microwindows in the  $\nu_3$ band provided coverage between altitudes of 5 and 25 km. Ten microwindows in the vs band were used to measure HCN between altitudes of 6 and 30 km, The profiles for SF, and CHCLF, were derived from unresolved Q branches on the basis of temperature and pressure dependent absorption coefficients. A single microwindow was fitted for each molecule, For  $SF_6$ , the microwindow spanned 945,0 to 952.0 cm<sup>-1</sup>, covering the intense  $\nu_a$  band Q branch between altitudes of 13 to 20 km, For CHClF<sub>2</sub>, a 0.5-cm<sup>-1</sup> wide microwindow centered at 829.05 cm-a was used measure the  $2\nu_8$  Q branch between 13 and 33 km. Examples of the target features in 1 ower stratospheric ATMOS

occultation spectra have been presented previously [Zander et al., 1987; 1988; Rinsland et al., 1993], Profiles of NaO are obtained by combining measurements from microwindows in several bands, depending on the filter selected for the occultation. Ground-based spectroscopic measurements indicate an average N<sub>2</sub>0 exponential rate of increase of (0.36±0.06)% yr<sup>-1</sup> between 1984 and 1992 [Zander et al., 1994a], Based on this result, the VMR changes for each molecule noted below have been multiplied by 0.966 to correct for the N20 increase between 1985 and 1994. Measurements with relative uncertainties larger than 20% for HCN, OCS, and SF<sub>8</sub> and 359 for CHC#F<sub>2</sub> have been excluded from the analysis.

#### Results

Carbonyl Sulfide (OCS)

Figure 1 presents the VMR ocs measurements plotted versus simultaneously measured N20 VMRs. The solid and dashed lines show OCS VMRs derived by averaging the measurements in intervals of 0,1 in log10(N20). The 1994-to-1985 OCS VMR ratio derived from these averages is 0.94 ± 0,03, 1 sigma. Assuming an exponential model for the OCS VMR increase with time, the corresponding trend is  $(-0.6\pm0.3)$ % yr<sup>-1</sup>. Hence, no significant change in the OCS VMR in the lower stratosphere occurred over this time period,

The inference of a negligible long. term OCS trend is consistent with the results obtained from high resolution ground-based IR measurements [Rinsland et al., 1992], Total columns retrieved from spectra recorded at the National Solar (NSO, latitude, 31.9°N Observatory 111.6°W longitude) between May 1977 and March 1991 showed a trend of (0.1±0.2) yr<sup>-1</sup>, 2 sigma, while similar □easurements at the International Station of the Jungfraujoch (ISSJ, 46.5°N latitude, 8.0°E longitude) between October 1984 and April 1991 yielded a trend of  $(-0.1\pm0.5)$ % yr<sup>-1</sup>, 2 sigma. Budget studies suggest that the main sources of OCS are natural

with biomass burning, the largest anthropogenic source, contributing about 10: to the total [Khalil and Rasmussen, 1984; Crutzen and Andreae, 1990], eruption of Mount Pinatubo (15.14°N, 120.35°E) in the Philippines on June 15, 1991, produced the largest volcanic input to the stratosphere ever observed by satellite instrument [McCormick, 1992], but as shown here, apparently no significant long-term change in lower stratospheric OCS resulted.

### Hydrogen Cyanide (HCN)

Figure 2 presents the stratospheric HCN VMRs plotted versus the simultaneous measurements of N2O VMRs, The format is the same as in Fig, 1. Based on the mean and standard deviation calculated from the averages shown by the solid and dashed lines, the 1994-to-1985 HCN VMR ratio is 1.09 ± 0.09, 1 sigma. This value corresponds to a exponential increase rate of (0.9±0.9) yr<sup>-1</sup>. Hence, there is marginal evidence for an increase in HCN VMRs In the lower stratosphere over the 94 year period separating the Spacelab 3 and ATLAS 3 missions.

To our knowledge, the only previous investigation of the HCN long-term trend was reported by Mahieu et al, [1995], In that study, high resolution ground-based IR measurements obtained at NSO between June 1984 and June 1993 and at ISSJ between May 1978 and July 1992 were analyzed to derive total vertical column abundance \*. Excluding the springtime measurements above both stations, which showed variable increases associated with elevated concentrations near the ground, HCN long-term column trends of (. J. 30±07. 1507 % yr 17 above NSO (0.99±0.51) yr<sup>-1</sup> above ISSJ were deduced. Hence, the present results are are in better agreement with the measurements [Mahieu et al., 1.995], which suggest a small, long-term increase in HCN amounts may have occurred,

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Sulfur hexafluorida (SF<sub>6</sub>)

The Spacelab 3 and ATLAS 3 lower stratospheric measurements of SF, near 30'N latitude are presented in Fig. 3. A substantial VMR increase over the 9½ years separating the 2 missionsis readily apparent, Based on the averages illustrated by the solid and dashed lines, the 1994-to-1985 SF, VMR ratio in the lower stratosphere is 1.95±0.06, 1 sigma, which corresponds to an exponential increase rate of (7.1±0.3)% yr<sup>-1</sup>.

The present result is in excellent agreement with 3 recent determination of the SFs long-term trend, all based on high resolution infrared solar absorption spectra, Rinsland et al. [1990] deduced exponential increase rate of (7,4\*1,9)% yr<sup>-1</sup> from comparison of solar occultation measurements recorded near 30°N latitude between March 1981 and June 1988 in the 12.18 km altitude region, Zander et al, [1991] reported SF<sub>6</sub> total column exponential increase rates of  $(6.6\pm7.2)$  yr<sup>-1</sup>, 2 sigma, above NSO between March 1981 and June 1990 and (6.9±2.8) yr<sup>-1</sup>, 2 sigma, above ISSJ between June 1986 and June 1990, The larger uncertainty in the NSO measurement result of the the increased difficulty in measuring SF, from a relatively low altitude site where strong H<sub>2</sub>C<sub>I</sub> and CO<sub>2</sub> interference overlap the target \$F, v, band Q branch (the NSO FTS is at an altitude of 2095m as compared to 3580m for the ISSJ FTS). An exponential increase rate of  $(8.7\pm2.2)$   $yr^{-1}$ , 2 sigma, was inferred by comparing University of Denver (32°N), ATMOS Spacelab 3 (31-N), ATMOS/ATLAS 1 (28°S, measurements in the 12-18 km altitude region after applying a small correction for the SF6 interhemispheric gradient based on Godel calculation [Rinsland et al., 1993],

Although the atmospheric concentration of  $SF_8$  is continuing to increase rapidly and the gas is likely to have a very long lifetime, its abundance is very low, The projected greenhouse warming potential of  $SF_8$  on a per molecule

basis is high, but the net warming due to  $SF_5$  will remain negligible with respect to that of  $CO_2$ , at least through 2010 [Ko et al., 1993].

### CHCLF2 (HCFC-22)

and ATLAS 3 The Spacelab 3 measurement comparison shown in Figure 4 shows the substantial buildup of hydrochlorofluorocarbon (HCFC)-22 that has occurred in the lower stratesphere in response to the increasing was, this molecule as a substitute for the 'GFGs, particularly CCl<sub>2</sub>F<sub>2</sub> (CFC-12) and CCl<sub>3</sub>F (CFC-11) [AFEAS, 1991a,b]. A 1994-to-1985 HCFC-22 lower stratospheric VMR ratio derived from these measurements is 2.08±0.24, 1 sigma, which corresponds to an exponential HCFC-22 increase rate of  $(7.7\pm1.2)$  yr<sup>-1</sup>.

The present result is in good agreement with recent IR and in situ HCFC-22 trend determinations, Zander et [1994] reported total column exponential increase rates of (7.010.23) yr above NSO from December 1980 to April 1992 and (7.0±0.35) yr<sup>-1</sup> above ISSJ from June 1986 to November 1992 while Irion et al. [1994] inferred an exponential total column increase rate of (6.7±0.5) yr<sup>-1</sup> on the basis of ATMOS IR solar spectra recorded from Mountain the Table Facility, Wrightwood, California (34.4°N), between October 1985 and July 1990, A slightly higher growth rate of  $(7.3\pm0.3)$  yr<sup>-1</sup> was deduced by Montzka et [1993] from surface ambient air mid-1987 sampling measurements  ${ t from}$ Solar occultation IR through 1992, measurements near 30°N latitude between March 1981 and June 1988 also yielded a higher exponential increase rate,  $(9.4\pm1.3)$ t yr<sup>-1</sup> in the 12-18 km altitude , region [Rinsland et al., 1990].

Empirically-based determinations/global ozone depletion potentials for HCFC-22 are as much as a factor of 2 larger than values from some gas phase models [Solomon et al., 1992], This result, coupled with the steady increase in atmospheric HCFC-22 concentrations indicated by recent measurements,

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suggests that HCFC-22 will continue to be ban important "contributor to depletion in the stratosphere.

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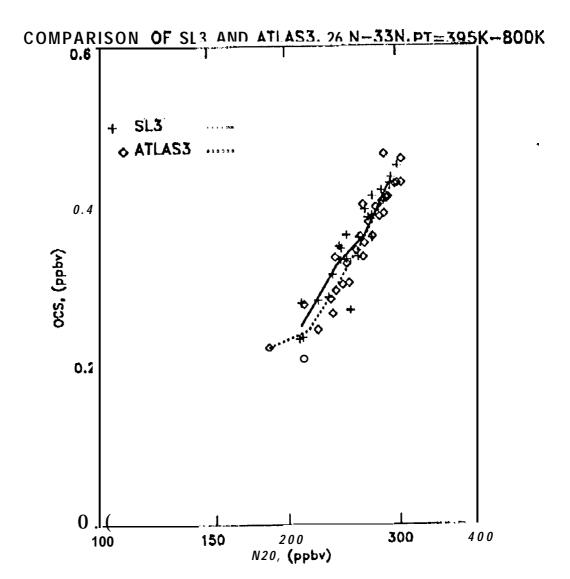
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## Figure Captions

- Fig. 1. ATMOS Spacelab 3 nd ATLAS 3 VMRs of OCS plotted versus simultaneously measured N<sub>2</sub>O VMRs (both in ppbv, 10-0), The measurements ware obtained between latitudes of 26\*N and 33°N and 0 between 395 K and 800 K (altitudes of ~17 to 30 km). Solid and dashed lines show average OCS VMRs calculated as a function of N<sub>2</sub>O from the 1985 and 1994 measurements, respectively, See text for details.
- Fig, 2. ATMOS Spacelab 3 and ATLAS 3 VMRs of HCN plotted versus simultaneous measured N20 VMRs (both in ppbv, lo-Q). The measurements were obtained between latitudes of 26"N and 33\*N and 0 between 395 K and 800 K (altitudes of ~17 to 30 km), Solid and dashed lines show average HCN VMRs calculated as a function of N20 from the 1985 and 1994 measurements, respectively. See text for details,
- Fig. 3, ATMOS Spacelab 3 and ATIAS 3 VMRs of SF<sub>6</sub> plotted versus simultaneous measured N<sub>2</sub>O VMRs (both in ppbv, 10<sup>-8</sup>). The measurements were obtained between latitudes of 26\*N and 33\*N and 0 between 395 K and 800 K (altitudes of ~17 to 30 km), Solid and dashed lines show average SF<sub>6</sub> VMRs calculated as a function of N<sub>2</sub>O from the 1985 and 1994 measurements, respectively. See text for details,
- Fig. 4. ATMOS Spacelab 3 and ATLAS 3 VMRs of CHCIF<sub>2</sub> (HCFC-22) plotted versus simultaneous measured N<sub>2</sub>O VMRs (both in ppbv, 10<sup>-9</sup>). The measurements were obtained between latitudes of 26°N and 33\*N and θ between 395 K and 800 K (altitudes of ~17 to 30 km). Solid and dashed lines show average HCFC-22 VMRs calculated as a function of N<sub>2</sub>O from the 1985 and 1994 measurements, respectively. See text. for details.
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Figure 1

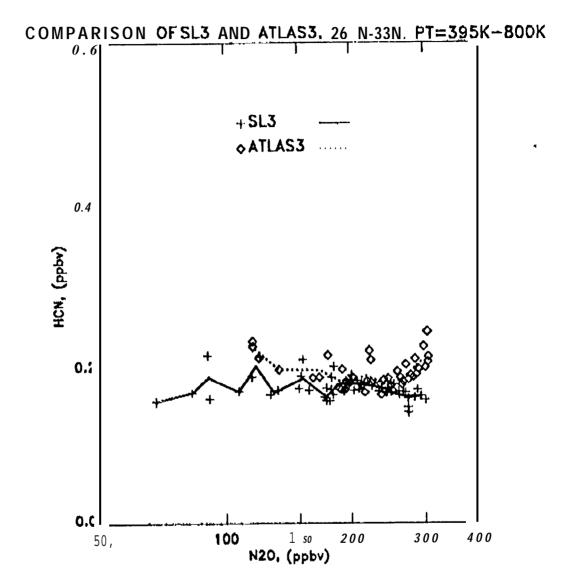


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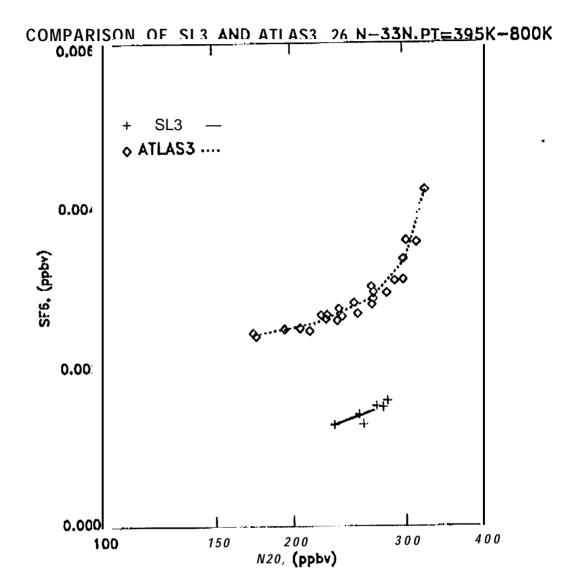
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Figure 3

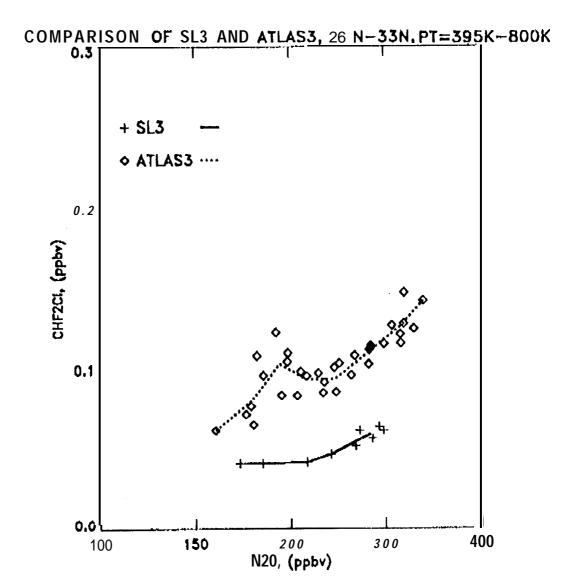


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# Figure 4



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